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## PATENT COOPERATION TREATY

From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

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01. Feb 2006

Erl. ....

# PCT

NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL PRELIMINARY  
REPORT ON PATENTABILITY

(PCT Rule 71.1)

Date of mailing  
(day/month/year)

03.02.2006

Applicant's or agent's file reference  
P60184PCT

**IMPORTANT NOTIFICATION**

International application No.  
PCT/EP2004/008492

International filing date (day/month/year)  
28.07.2004

Priority date (day/month/year)  
08.08.2003

Applicant

ATOTECH DEUTSCHLAND GMBH

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary report on patentability and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

#### 4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the International preliminary report on patentability. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed inventions is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability, requirements for enabling disclosure, clarity and support for the claims.

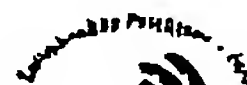
# PATENT COOPERATION TREATY

## PCT

### INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference <b>P60194PCT</b>	<b>FOR FURTHER ACTION</b> See Form PCT/PEA/416	
International application No. <b>PCT/EP2004/008492</b>	International filing date (day/month/year) <b>28.07.2004</b>	Priority date (day/month/year) <b>08.08.2003</b>
International Patent Classification (IPC) or national classification and IPC <b>C25D3/08</b>		
Applicant <b>ATOTECH DEUTSCHLAND GMBH</b>		
<p>1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 8 sheets, including this cover sheet.</p> <p>3. This report is also accompanied by ANNEXES, comprising:</p> <p>a. <input checked="" type="checkbox"/> sent to the applicant and to the International Bureau) a total of 6 sheets, as follows:</p> <p><input checked="" type="checkbox"/> sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).</p> <p><input type="checkbox"/> sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.</p> <p>b. <input type="checkbox"/> (sent to the International Bureau only) a total of (indicate type and number of electronic carrier(s)) . containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).</p>		
<p>4. This report contains indications relating to the following items:</p> <p><input checked="" type="checkbox"/> Box No. I Basis of the opinion</p> <p><input type="checkbox"/> Box No. II Priority</p> <p><input checked="" type="checkbox"/> Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</p> <p><input type="checkbox"/> Box No. IV Lack of unity of invention</p> <p><input checked="" type="checkbox"/> Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</p> <p><input type="checkbox"/> Box No. VI Certain documents cited</p> <p><input type="checkbox"/> Box No. VII Certain defects in the international application</p> <p><input checked="" type="checkbox"/> Box No. VIII Certain observations on the international application</p>		
Date of submission of the demand  <b>20.06.2005</b>	Date of completion of this report  <b>03.02.2006</b>	
Name and mailing address of the international preliminary examining authority:  European Patent Office - P.O. Box 5818 Patentplatz 2	Authorized Officer  	

**10/566913**

**IAP20 Rec'd PCT/PTO 03 FEB 2006**

**INTERNATIONAL PRELIMINARY REPORT  
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**Box No. I Basis of the report**

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.
  - ☐ This report is based on translations from the original language into the following language, which is the language of a translation furnished for the purposes of:
    - ☐ international search (under Rules 12.3 and 23.1(b))
    - ☐ publication of the international application (under Rule 12.4)
    - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)
2. With regard to the **elements\*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

**Description, Pages**

1-17 as originally filed

**Claims, Numbers**

1-23 received on 21.06.2005 with letter of 20.06.2005

- ☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing

3. ☒ The amendments have resulted in the cancellation of:

- ☐ the description, pages
- ☒ the claims, Nos. 24
- ☐ the drawings, sheets/figs
- ☐ the sequence listing (*specify*):
- ☐ any table(s) related to sequence listing (*specify*):

4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

- ☐ the description, pages
- ☐ the claims, Nos.
- ☐ the drawings, sheets/figs
- ☐ the sequence listing (*specify*):
- ☐ any table(s) related to sequence listing (*specify*):

\* If item 4 applies, some or all of these sheets may be marked "superseded."

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### Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:
- ☐ the entire international application,
  - ☒ claims Nos. 1, 20, 23 (all partially)  
because:
    - ☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (specify):
    - ☐ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. are so unclear that no meaningful opinion could be formed (*specify*):
    - ☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.
    - ☒ no international search report has been established for the said claims Nos. 1, 20, 23 (all partially)
    - ☐ the nucleotide and/or amino acid sequence listing does not comply with the standard provided for in Annex C of the Administrative Instructions in that:
      - the written form ☐ has not been furnished
      - ☐ does not comply with the standard
      - the computer readable form ☐ has not been furnished
      - ☐ does not comply with the standard
    - ☐ the tables related to the nucleotide and/or amino acid sequence listing, if in computer readable form only, do not comply with the technical requirements provided for in Annex C-bis of the Administrative Instructions.
  - ☐ See separate sheet for further details

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### Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

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#### 1. Statement

Novelty (N)	Yes: Claims	1-23
	No: Claims	
Inventive step (IS)	Yes: Claims	1-23
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-23
	No: Claims	

#### 2. Citations and explanations (Rule 70.7):

see separate sheet

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### Box No. VIII Certain observations on the international application

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The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet



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**PCT/EP2004/008492**

**Re Item I**

**Basis of the report**

Amended claims as submitted with letter of 21/06/05

**Re Item III**

**Non-establishment of opinion with regard to novelty, inventive step and industrial applicability**

Claims 1, 20 and 23 are not supported by the description as required by Article 6 PCT, as their scope is broader than justified by the description. The reasons therefor are the following: present claims 1, 20 and 23 relate to an extremely large number of possible compounds. Support within the meaning of Article 6 PCT and/or disclosure within the meaning of Article 5 PCT is to be found, however, for only a very small proportion of the compounds claimed. Hence, the examples shown in the description describe copper plating solutions containing a specific aromatic halogen derivative (either aldehyde or hydroxy substituted). Substituents of the acetyl group and hydroxyalkyl group would be considered to behave similarly to those exemplified compounds and thus supported by the description.

The claims however also encompass aromatic halogen derivatives substituted with hydrogen only as well as aromatic halogen derivatives substituted with alkyl group only. It is however clear from the description on page 6 lines 15-20 that an hydroxy group either present in the compound or upon reaction with the solution is the active substance. Therefore, generalisation of the examples compounds to these compounds are not supported by the description.

**Re Item V**

**Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

- 1 The following documents are referred to in this communication:  
D1: EP 1 300 486 A (SHIPLEY CO. L.L.C) 9 April 2003  
D2: EP 1 300 487 A (SHIPLEY CO. L.L.C) 9 April 2003  
D3: FR-A-2 139 724 (LABOLAC,FR) 12 January 1973

**2 Citations**

Document D1 discloses: (the references in parentheses applying to this document): an

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aqueous acidic solution for electrolytically depositing copper coatings (claim 14-23, 33-42), said solution containing at least one oxygen-containing, high molecular additive (page 6 l. 35-39), at least one water soluble sulfur compound (page 6 l. 27-34) and at least one aromatic halogen derivative ("2-chloro-4-hydroxybenzaldehyde"), said derivative being used at a level of 0.01g/l to 20.0g/l (page 5 l. 27-47, example 2). The solution of D1 is suitable for depositing copper onto printed circuit board material as well as for producing copper coating in vertical and/or horizontal conveyORIZED plating lines (page 8 l. 15-17).

Document D2 discloses (the references in parentheses applying to this document): an aqueous acidic solution for electrolytically depositing copper coatings (claim 18, 23, 26, 28-31 and 46), said solution containing at least one oxygen-containing, high molecular additive (page 7 l. 35-39), at least one water soluble sulfur compound (page 7 l. 24-34) and at least one aromatic halogen derivative ("4-chloro-resorcinol, 3-chlorophenol"), said derivative being used at a level of 0.01g/l to 20.0g/l (page 6 l. 46-50, examples 2 and 3). The solution of D1 is suitable for depositing copper onto printed circuit board material as well as for producing copper coating in vertical and/or horizontal conveyORIZED plating lines (page 9 l. 15-17).

Document D3 discloses (the references in parentheses applying to this document): an aqueous acidic solution for electrolytically depositing bright metal coatings (Abstract), said solution containing halogen substituted phenol ("I to V, VII and VIII") in an amount of 12 to 20g/l (Claims 1 and 2).

**3 Novelty (Article 33(2) PCT)**

As can be seen from the above, none of the cited art discloses acidic copper solutions with a level of aromatic halogen derivative of 0.005mg/l to 0.9mg/l. The subject-matter of claims 1-23 is therefore new (Article 33(2) PCT).

**4 Inventive step (Article 33(3) PCT)**

The problem addressed by the present application is to provide copper solutions which achieve high levelling without compromising the bright appearance of the surface layer (page 4 l. 1-20). This is solved by using levels ranging from 0.005mg/l to 0.9mg/l of selected aromatic halogen derivative.

D1, considered as the closest prior art also discloses this problem (page 2 l. 49-50) and

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provides the use of substituted aldehyde compounds, in particular aromatic halogen derivative at a level of 0.01g/l to 20g/l (page 5 line 47), 0.1g/l (100mg/l) being exemplified (Ex.2).

The subject matter of claim 1 differs from that of D1 in that the substituted aromatic halogen are present at a low level. The problem is thus to find copper solution providing better deposition.

The solution to this problem proposed in claim 1 of the present application is considered as involving an inventive step (Article 33(3) PCT) for the following reasons:  
the solution to use low levels of specific aromatic are neither disclosed nor taught in the cited art, let alone for providing better bright metal surface.

The same reasoning applies mutatis mutandis to independent claims 20, 21 and 23 relating to the use of said solutions.

Claims 2-19 and 22 are dependent claims and as such also meet the requirements of the PCT with respect to novelty and inventive step.

**Re Item VIII**

**Certain observations on the International application**

The application does not meet the requirements of Article 6 PCT, because claim 1 is not clear.

The term "oxygen-containing, high molecular additive" used in claim 1 is vague and unclear and leaves the reader in doubt as to the meaning of the technical feature to which it refers, thereby rendering the definition of the subject-matter of said claim unclear, Article 6 PCT. The fact that this term is used in D1 and D2, both citations being in the name of the same Applicant, does not imply that the term is generally known.

As per mentioned in point III above, it is clear from the description on page 6 lines 15-20 that an hydroxy group either present in the compound or upon reaction with the solution is the active substance. Since some compounds falling within the definition of independent claim 1 do not contain this feature it does not meet the requirement following from Article 6 PCT taken in combination with Rule 6.3(b) PCT that any independent claim must contain all the technical



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features essential to the definition of the invention.

The vague and imprecise statement in the description on page 17 l. 28-34 implies that the subject-matter for which protection is sought may be different to that defined by the claims, thereby resulting in lack of clarity (Article 6 PCT) when used to interpret them.

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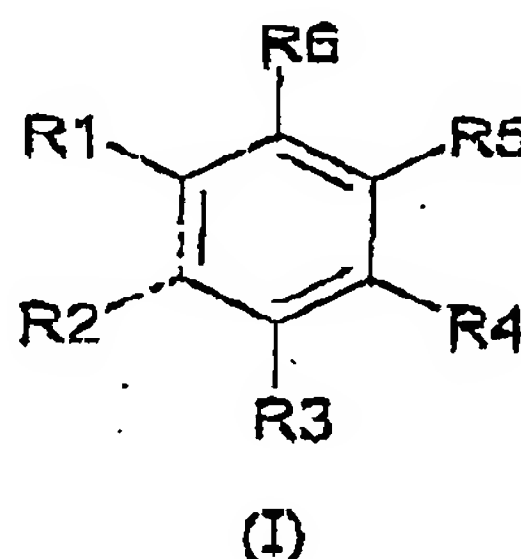
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21. 06. 2005

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# Claims

1. An aqueous acidic solution for electrolytically depositing copper coatings, said solution containing at least one oxygen-containing, high molecular additive  
5 and at least one water soluble sulfur compound, characterized in that the solution additionally contains at least one aromatic halogen derivative having the general formula (I)



10

wherein

- R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub>, R<sub>5</sub> and R<sub>6</sub> are each independently radicals selected from the  
15 group comprising hydrogen, aldehyde, acetyl, hydroxy, hydroxyalkyl having 1 – 4 carbon atoms, alkyl having 1 – 4 carbon atoms and halogen, with the proviso that the number of radicals R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub>, R<sub>5</sub> and R<sub>6</sub> which are halogen ranges from 1 – 5, wherein

- 20 ~~12.~~ The solution according to claim 1, characterized in that the concentration of the at least one aromatic halogen derivative ranges from about 0.005 – about 0.9 mg/l.

- ~~2~~  
~~2.~~ The solution according to any one of the preceding claims, characterized  
25 in that the aldehyde is selected from the group comprising formyl (–CHO), methylformyl (–CH<sub>2</sub>–CHO) and ethylformyl (–C<sub>2</sub>H<sub>4</sub>–CHO).

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- 3  
4. The solution according to any one of the preceding claims, characterized in that alkyl is branched or unbranched and is selected from the group comprising methyl, ethyl, *n*-propyl, *iso*-propyl, *n*-butyl, *iso*-butyl and *tert*-butyl.
- 5  
4  
5. The solution according to any one of the preceding claims, characterized in that alkyl is hydroxyalkyl and that it is branched or unbranched.
- 5  
5. The solution according to any one of the preceding claims, characterized in that at least one hydroxyalkyl is hydroxymethyl.
- 10  
6  
7. The solution according to any one of the preceding claims, characterized in that the at least one aromatic halogen derivative is selected from the group comprising
- 15  
2-chlorobenzaldehyde  
2-chlorophenol  
4-chloro-3-methylphenol  
2-chloro-4,5-dimethylphenol  
20 4-chloro-3,5-dimethylphenol  
4-chlorophenol  
3-chlorophenol  
o-chloroacetophenone  
2-chlorobenzyl alcohol  
25 4-bromo-2,6-dimethylphenol  
4-bromophenol  
2,4-dichlorobenzyl alcohol  
2,6-dibromo-4-methylphenol  
2,5-dichlorophenol  
30 3,5-dibromobenzaldehyde  
2,5-dibromobenzoic acid  
2,4,6-trichlorophenol  
2,3,6-trichlorobenzaldehyde.

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~~7~~

~~8.~~ The solution according to any one of the preceding claims, characterized in that the at least one oxygen-containing, high molecular additive is selected from the group comprising

5

polyvinyl alcohol  
carboxymethyl cellulose  
polyethylene glycol  
polypropylene glycol  
stearic acid polyglycol ester

10

oleic acid polyglycol ester  
stearyl alcohol polyglycol ether  
nonylphenol-polyglycol ether  
octanol polyalkylene glycol ether  
octanediol-bis-(polyalkylene glycol ether)

15

poly(ethylene glycol-*ran*-propylene glycol)  
poly(ethylene glycol)-*block*-poly(propylene glycol)-*block*-poly(ethylene glycol) and  
poly(propylene glycol)-*block*-poly(ethylene glycol)-*block*-poly(propylene glycol).

20

~~8~~

~~9.~~ The solution according to any one of the preceding claims, characterized in that the at least one water soluble sulfur compound is selected from the group comprising organic, nitrogen-free thio compounds and the salts thereof.

25

~~9~~

~~10.~~ The solution according to claim ~~8~~, characterized in that the salts contain alkali or earth alkali metal ions, selected from the group comprising sodium, potassium, magnesium and calcium

30

~~10~~

~~11.~~ The solution according to any one of claims ~~8~~ and ~~10~~, characterized in that the at least one organic nitrogen-free thio compound is selected from the group comprising

sodium salt of 3-(benzthiazolyl-2-thio)-propylsulfonic acid  
sodium salt of 3-mercapto propane-1-sulfonic acid

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- disodium salt of thiophosphoric acid-O-ethyl-bis-( $\omega$ -sulfopropyl)-ester  
trisodium salt of thiophosphoric acid-tris-( $\omega$ -sulfopropyl)-ester  
sodium salt of ethylenedithio dipropyl sulfonic acid  
disodium salt of bis-( $p$ -sulfophenyl)-disulfide  
5 disodium salt of bis-( $\omega$ -sulfopropyl)-sulfide  
disodium salt of bis-( $\omega$ -sulfopropyl)-disulfide  
disodium salt of bis-( $\omega$ -sulfohydroxypropyl)-disulfide  
disodium salt of bis-( $\omega$ -sulfobutyl)-disulfide  
sodium salt of methyl-( $\omega$ -sulfopropyl)-disulfide  
10 sodium salt of methyl-( $\omega$ -sulfobutyl)-trisulfide  
potassium salt of O-ethyl-dithiocarbonic acid-S-( $\omega$ -sulfopropyl)-ester  
thioglycolic acid

- <sup>11</sup>  
12. The solution according to any one of the preceding claims, characterized  
15 in that acid is contained in the solution and that the acid is selected from the  
group comprising sulfuric acid, hydrochloric acid, fluoboric acid and  
methanesulfonic acid.

- <sup>12</sup>  
13. The solution according to any one of the preceding claims, characterized  
20 in that the solution additionally contains chloride ions.

- <sup>11</sup> <sup>12</sup>  
14. The solution according to claim 13, characterized in that the chloride ions  
have been added to the solution in the form of sodium chloride and/or of  
hydrochloric acid.

- 25 <sup>14</sup>  
15. The solution according to any one of the preceding claims, characterized  
in that the solution additionally contains at least one organic, nitrogen-  
containing thio compound.

- <sup>15</sup> <sup>14</sup>  
30 16. The solution according to claim 15, characterized in that the at least one  
nitrogen-containing thio compound is selected from the group comprising

thiourea

N-acetylthiourea



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- 5 N-trifluoroacetyl thiourea  
N-ethylthiourea  
N-cyanoacetyl thiourea  
N-allylthiourea  
o-tolylthiourea  
N,N'-butylene thiourea  
thiazolidine thiol-2  
4-thiazoline thiol-2  
imidazolidine thiol-2-(N,N'-ethylene thiourea)  
10 4-methyl-2-pyrimidine thiol  
2-thiouracil

- 16  
17. The solution according to any one of the preceding claims, characterized  
in that the solution additionally contains at least one polymeric phenazinium  
15 compound.

- 17 16  
18. The solution according to claim 17, characterized in that the at least one  
polymeric phenazinium compound is selected from the group comprising

- 20 poly(6-methyl-7-dimethylamino-5-phenyl-phenazinium sulfate)  
poly(2-methyl-7-diethylamino-5-phenyl-phenazinium chloride)  
poly(2-methyl-7-dimethylamino-5-phenyl-phenazinium sulfate)  
poly(5-methyl-7-dimethylamino-phenazinium acetate)  
poly(2-methyl-7-anilino-5-phenyl-phenazinium sulfate)  
25 poly(2-methyl-7-dimethylamino-phenazinium sulfate)  
poly(7-methylamino-5-phenyl-phenazinium acetate)  
poly(7-ethylamino-2,5-diphenyl-phenazinium chloride)  
poly(2,8-dimethyl-7-diethylamino-5-p-tolyl-phenazinium chloride)  
poly(2,5,8-triphenyl-7-dimethylamino-phenazinium sulfate)  
30 poly(2,8-dimethyl-7-amino-5-phenyl-phenazinium sulfate)  
poly(7-dimethylamino-5-phenyl-phenazinium chloride)

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- <sup>18</sup>  
~~18~~ The solution according to any one of the preceding claims, characterized in that the solution additionally contains at least one polymeric nitrogen compound.
- 5 <sup>19</sup>  
~~20~~ The solution according to claim <sup>18</sup>~~19~~, characterized in that the at least one polymeric nitrogen compound is selected from the group comprising polyethylene imine, polyethylene imide, polyacrylic acid amide, polypropylene imine, polybutylene imine, N-methyl polyethylene imine, N-acetyl polyethylene imine, N-butyl polyethylene imine.
- 10 <sup>20</sup>  
~~21~~ Use of the solution according to any one of claims 1 - <sup>19</sup>~~20~~ for depositing a copper coating.
- <sup>21</sup>  
~~22~~ Use of the solution according to ~~any one of claims 1 - 20~~ <sup>20</sup>for depositing
- 15 copper onto printed circuit board material.
- <sup>22</sup>  
~~23~~ Use according to any one of claims <sup>20</sup>~~21~~ and <sup>21</sup>~~22~~ for producing copper coatings in vertical and/or horizontal conveyORIZED plating lines.
- 20 <sup>23</sup>  
~~24~~ A method of electrolytically depositing copper coatings on metal or plastic surfaces, comprising bringing the surfaces into contact with the solution according to any one of claims 1 - <sup>19</sup>~~20~~ and electrolytically depositing copper onto the surfaces.

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